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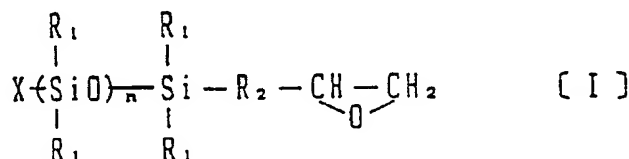
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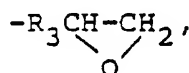
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(54) Thermoplastic resin composition.

⑤7) A thermoplastic resin composition comprises a thermoplastic resin (A) having a solubility parameter δ of at least $9.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ in which a modifier (B) having a solubility parameter δ of not higher than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ is dispersed to form a dispersion, characterized by containing a dispersibility improver (C) comprising a silicon-containing polyacetal copolymer prepared by bonding 0.01 to 30 wt.% of a terminal-reactive silicone oil of the following general formula [I]:



wherein R₁ represents an alkyl or phenyl group, R₂ represents a divalent organic group, X represents a group selected from the group consisting of -NH₂, -OH,



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$-R_4OH$, $-R_5COOH$, $-R_6COOR_7$, $-R_8$ and $-OR_9$, R_3 represents a divalent organic group, R_4 to R_6 each represent an alkylene group having 1 to 10 carbon atoms, R_7 to R_9 each represent an alkyl group having 1 to 10 carbon atoms and n represents a number of 5 to 10.

THERMOPLASTIC RESIN COMPOSITION

The present invention relates to a synthetic resin, particularly a thermoplastic resin. More particularly, the present invention relates to a composite thermoplastic resin composition having a high toughness and an excellent abrasion resistance in which a modifier which is not substantially homogeneously dispersible is homogeneously dispersed in the presence of a siloxane copolymer as a dispersibility improver.

[Prior Art]

Engineering plastics such as polyacetal resins; aromatic thermoplastic polyesters such as polyethylene terephthalate and polybutylene terephthalate; thermoplastic polyamides such as nylon 6 and nylon 6,6; and polyesteramides which are copolymers of the abovementioned polymers are widely used as resins having excellent tensile strength, tear strength, resilience, cold resistance and bending resistance. Attempts have been made to add various substances to the resin to form a composition having modified properties and improved functions. It is known that when a solid filler is to be used, coupling agents and some surfactants are also used in order to improve the compatibility of the surface of the filler with the matrix resin or to obtain a homogeneous dispersion.

However, when the modifiers are resins or liquid substances, they have solubility parameters δ which are different from one another and, therefore, the compatibility of them with one another is so poor that no homogeneous dispersion can be obtained in many cases.

In such a case, the above-described coupling agents are useless and ordinary surfactants have a poor heat stability and are unsuitable for the resins called engineering plastics which necessitate the processing at a high temperature. Particularly when an oily substance or the like is to be dispersed, it cannot be dispersed in a large amount and is separated to cause bleeding. Even when it is used in a small amount, no homogeneous dispersion can be obtained, the appearance of the product is impaired, the physical properties thereof are not uniform and mechanical strengths thereof tend to be reduced. Also when the polymers are blended together, no uniform dispersion can be easily obtained as in the dispersion of the oily substance, the surface peeling phenomenon is caused and no uniform mechanical strengths can be easily obtained.

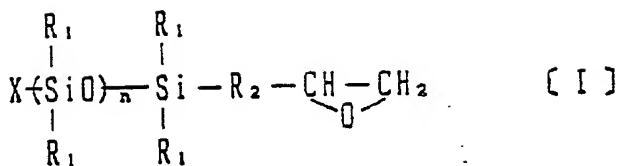
Processes for solving these problems have been proposed. They include, for example, a process wherein a block copolymer having a segment which is the same as that of the substance to be incorporated therein is used and a process wherein the substances to be incorporated are bonded together by, for example, an exchange reaction on the surface thereof to improve the surface affinity. However, these processes lack versatility, because the substances to be incorporated are limited, other substances cannot be treated by these processes and different formulations are necessary depending on the substances to be incorporated.

[Summary of the Invention]

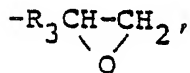
An object of the present invention is to provide a thermoplastic resin composition having a high toughness and excellent abrasion resistance, which is prepared by melt-blending to obtain a homogeneous dispersion without causing bleeding or reduction in mechanical and physical properties.

After intensive investigations made under these circumstances, the inventors have found that specified siloxane copolymers are not decomposed even at a high temperature, that they have such a surface-activating effect that they can be used widely for various materials to be incorporated and that they are capable of forming a homogeneous dispersion even with a substance having a solubility parameter far different from that of them. The present invention has been completed on the basis of these findings.

The present invention relates to a thermoplastic resin composition comprising a thermoplastic resin (A) having a solubility parameter δ of at least $9.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ in which a modifier (B) having a solubility parameter δ of not higher than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ is dispersed to form a dispersion, characterized by containing a dispersibility improver (C) comprising a silicon-containing polyacetal copolymer prepared by bonding 0.01 to 30 wt.% of a terminal-reactive silicone oil of the following general formula [I]:



wherein R₁ represents an alkyl or phenyl group, R₂ represents a divalent organic group, X represents a group selected from the group consisting of -NH₂, -OH,



-R₄OH, -R₅COOH, -R₆COOR₇, -R₈ and -OR₉, R₃ represents a divalent organic group, R₄ to R₆ each represent an alkylene group having 1 to 10 carbon atoms, R₇ to R₉ each represent an alkyl group having 1 to 10 carbon atoms and n represents a number of 5 to 10,

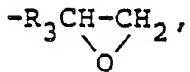
with 99.99 to 70 wt.% of a polyacetal segment through a covalent bond, the degree of polymerization of the siloxane segment (n in the formula [I]) being 5 to 1,000 and the degree of polymerization of the polyacetal segment being 5 to 2,000, in which the weight ratio of (A) to (B) is 99.9 to 85/0.1 to 15 and the amount of (C) is 0.01 to 5 parts by weight for 100 parts by weight in total of (A) and (B).

The polyacetal segments (C) herein comprise polyoxymethylene units as the main skeleton. They are homopolymers formed by polymerizing formaldehyde or trioxane or copolymers comprising formaldehyde or trioxane as the main component and one or more comonomers selected from among cyclic ethers and cyclic formals such as ethylene oxide, propylene oxide, oxetane, dioxolane, dioxepane, trioxepane and formal of butanediol.

The silicon-containing polyacetal copolymers (C) are prepared by a known polyacetal polymerization process such as a process wherein a terminal-reactive silicone oil of the general formula [I] in an amount of at least 0.01 wt.% based on the total reactive monomers is copolymerized with formaldehyde or trioxane under heating in the presence of a catalyst; or a process wherein formaldehyde or trioxane as the main component is copolymerized with one or more comonomers selected from the group consisting of cyclic ethers and cyclic formals such as ethylene oxide, propylene oxide, oxetane, dioxolane, dioxepane, trioxepane and formal of butanediol and the terminal-reactive silicone oil of the general formula [I] under heating in the presence of a catalyst. Paraformaldehyde can also be used as the material for feeding formaldehyde.

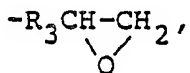
The terminal-reactive silicone oils used in the present invention are those represented by the above general formula [I]. They are used either alone or in combination of two or more of them.

When X in the above general formula [I] is a group other than



the copolymer obtained is a block copolymer or a comblike graft copolymer.

When X is



namely when the silicone oil has glycidyl groups at both ends, a graft copolymer comprising polyacetal segments crosslinked with a siloxane segment is obtained. A particularly preferred silicone oil is a polydimethylsiloxane diglycidyl ether of the general formula [I] wherein R₁ represents a methyl group and R₂ and R₃ each represent -CH₂-, -OCH₂-, -CH₂CH₂CH₂OCH₂- or the like which has a degree of polymerization of 5 to 1,000, preferably 10 to 500.

In the formula [I], it is preferable that R_1 represents an alkyl group with 1 to 4 carbon atoms, especially 1 or 2, most preferably methyl; R_2 and R_3 each represent a group of the formula $-R_{10}-Y-R_{11}$ in which R_{10} and R_{11} each represent an alkyl group with 1 to 6 carbon atoms and Y represents oxygen or sulphur, or R_2 and R_3 each represent an alkylene group with 1 to 12 carbon atoms or an oxyalkylene group with 1 to 12 carbon atoms.

The weight ratio of the above-described terminal-reactive silicone oil to the monomer constituting the polyacetal segment is 0.01 to 30/99.99 to 70, preferably 0.02 to 20/99.98 to 80.

The silicon-containing polyacetal copolymer (C) of the present invention is produced under the same conditions as those of known processes for producing polyacetals in the presence of the same catalyst as that of the latter processes.

In a particularly preferred embodiment of the process, trioxane as the main monomer is reacted with a small amount of a cyclic ether or a cyclic formal as the comonomer and polydimethylsiloxane diglycidyl ether in the presence of a cationic active catalyst such as boron trifluoride or a complex compound thereof at 60 to 150 °C.

A known polymerization regulator such as dimethyl acetal can be used in the production of the silicon-containing polyacetal copolymer (C) within the scope of the present invention.

As the measure of the evaluation of the compatibility of the polymers, the solubility parameter δ is widely employed. This method is described in detail in many books (see, for example, "Polymer Handbook", the Second Edition, IV, page 349).

As is well known, δ represents a cohesive strength of molecules of the same compound. In a mixture which satisfies the following formula [II]:

$$|\delta_1 - \delta_2| \approx 0 \quad \text{[II]}$$

wherein δ_1 and δ_2 represent solubility parameters of different substances from each other, swelling or dissolution occurs when the mixture comprises a combination of a high-molecular compound and a low-molecular compound, and a stable dispersion is kept by mechanical stirring when the mixture comprises a combination of high molecular compounds with each other. In both mixtures, the affinity is strong and any phase separation phenomenon scarcely occurs. When the value of the formula [II] exceeds 1, usually a phase separation phenomenon occurs so that no stable, homogeneous dispersion can be easily obtained.

An object of the present invention is to make it possible to effectively prepare a homogeneous dispersion comprising at least a substance (A) having δ of at least $9.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ with a substance (B) having δ not higher than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$.

The components (A) having δ of at least $9.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ used in the present invention are thermoplastic resins selected from the group consisting of, for example, polyesters, polyamides, polyesteramides, polyurethanes, polyvinyl chloride, polycarbonates, polyvinylidene chloride, polyacrylonitrile, polyacetals, polymethacrylonitrile, polymethyl acrylate, polyvinyl alcohol, celluloses and derivatives of them. They may be used either singly or in combination of two or more of them.

Among them, preferred are polyacetals, polyesters, polyamides, polyesteramides and polyurethanes.

Among them, most preferred are homopolymers and copolymers comprising polyoxymethylene units as the main skeleton.

The modifiers (B) having a solubility parameter δ of not higher than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ are one or more homopolymers selected from the group consisting of polytetrafluoroethylene, polyvinylidene chloride, polychlorotrifluoroethylene, fluororubber, polydimethylsilicone, polyalkylsiloxanes, polyphenylsiloxane, silicone rubber, butyl rubber, polyisobutylene, polyethylene, ethylene/propylene rubber and polypropylene and/or a copolymer comprising at least one of the monomer units forming these polymers as the constituent. The modifiers (B) are preferably fluorocarbon oil, silicone oil and mineral oil having a molecular weight of not higher than 200,000.

The ratios of the components of the composition of the present invention are as follows: the weight ratio of (A) to (B) is not particularly limited so far as the component (B) is substantially present even in a very small amount. Usually the ratio of (A) to (B) is 99.9 to 85/0.1 to 15. The amount of the component (C) is 0.01 to 5 parts by weight, particularly preferably 0.1 to 2 parts by weight, for 100 parts by weight in total of components (A) and (B).

When the amount of the component (B) is less than the above-described range, the characteristic features of the modifier cannot be easily obtained and, on the contrary, when it exceeds that range, the physical properties of the resin component (A) are excessively modified to make the homogeneous

dispersion thereof difficult unfavorably. When the amount of the component (C) is insufficient, no satisfactory homogeneous dispersibility can be obtained.

The resin composition of the present invention can be homogeneously mixed and dispersed with an ordinary apparatus for dissolving and kneading thermoplastic resins, such as an extruder, and it can be easily molded with a molding machine by an ordinary method.

The composition of the present invention is produced by mixing a synthetic resin with another substance having a δ ($\text{cal}^{1/2}\text{cm}^{-3/2}$) value different from that of the synthetic resin and therefore difficultly dispersible in the resin, in the presence of a special siloxane copolymer (C) to form a homogeneous dispersion. By this process, compounds having a δ value of less than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ and substantially incompatible with the base polymer can be quite easily dispersed and the bleeding in the course of the molding or after the molding can be prevented. Further by this process, high toughness and abrasion resistance can be imparted to the moldings without causing any surface peeling phenomenon. The thermoplastic resin composition of the present invention thus having excellent properties can be used for various purposes. Namely, they are usable as structural materials such as outside plates of automobiles of which excellent physical and mechanical properties such as impact resistance are required and as materials for parts of precision instruments such as key tops and gears as well as electric components of which excellent abrasion resistance and sliding resistance are required.

20 [Examples]

The following Examples will further illustrate the present invention. Referential Examples of the processes for producing the Polymer (C) containing a siloxane segment will be also given.

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Referential Example 1

0.02 part by weight of polydimethylsiloxane diglycidyl ether [number-average degree of polymerization: about 25] having glycidyl groups at both ends as the terminal-reactive silicone oil was added to a mixture of 98 parts by weight of trioxane with 2 parts by weight of ethylene oxide. The mixture was heated to 65°C under thorough stirring. Then a given amount of boron trifluoride/ethyl ether complex was added thereto and the polymerization was conducted at 60 to 100°C .

As the polymerization proceeded, the reaction system became milky and solidified. After 40 minutes, an aqueous alcohol solution containing a small amount of triethylamine was added thereto to terminate the reaction. The product was pulverized and thrown into acetone containing a small amount of triethylamine to wash it thoroughly. The product was air-dried and stabilized in the same manner as that employed in the production of the polyacetal resin.

40 Referential Example 2

The same procedure as that of Referential Example 1 was repeated except that 3 parts by weight of polydimethylsiloxane diglycidyl ether [number-average degree of polymerization: about 50] having glycidyl groups at both ends as the terminal-reactive silicone oil was added to a mixture of 98 parts by weight of trioxane with 2 parts by weight of ethylene oxide to prepare a silicon-containing polyacetal copolymer.

Referential Example 3

50 The same procedure as that of Referential Example 1 or 2 was repeated except that 3 parts by weight of polydimethylsiloxane monoglycidyl ether [number-average degree of polymerization: about 500] having a methyl group at an end (X) as the terminal-reactive silicone oil was added to a mixture of 98 parts by weight of trioxane with 2 parts by weight of ethylene oxide to give a silicon-containing polyacetal copolymer.

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Referential Example 4

The same procedure as that of Referential Example 3 was repeated except that 97 parts by weight of trioxane, 3 parts by weight of dioxolane and 10 parts by weight of the same silicone oil as that used in Referential Example 3 were used to give a silicon-containing polyacetal copolymer.

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Referential Example 5

The same procedure as that of the above-described Referential Examples was repeated except that 97 parts by weight of trioxane, 3 parts by weight of dioxolane and 20 parts by weight of polydimethylsiloxane monoglycidyl ether [number-average degree of polymerization: about 300] having -OCH₃ at an end (X) as the terminal-reactive silicone oil were used to give a silicon-containing polyacetal copolymer.

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Example 1 and Comparative Example 1

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A polyacetal resin (POM) (Duracon M90; a product of Polyplastics Co.) as the component (A), a silicone oil (SH-200; a product of Toray Silicone Co.) as the component (B) and a silicon-containing polyacetal copolymer prepared in Referential Example 1, 2 or 3 as the component (C) were kneaded together each in an amount shown in the following table with an extruder at 220°C. The composition was thus pelletized. The pellets were freeze-broken with liquid nitrogen. The fracture of the obtained resin composition was observed with a scanning electron microscope to find out that the silicon oil was homogeneously dispersed. The resin composition was injection-molded to form test pieces. After leaving the test pieces to stand at 80°C for one month, no bleeding of the oil was substantially observed on the surface. The coefficient of dynamic friction was determined according to ASTM D 1894. The test pieces having a size of 100 x 25 x 3 mm were repeatedly bent at an angle of 90° at room temperature according to JIS Z-2248, the internal bending radius being 1 mm. Even after repeating the bending 24 times, no delamination was observed on the bent surface. The same test pieces as above were subjected to extraction with acetone at room temperature for 1 h. The results are shown in the following table.

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The same composition as that of Example 1 except that no component (C) was contained was prepared and the test pieces thereof were subjected to the same test as above. The bleeding of the oil was recognized on the surfaces of the test pieces. The results are also shown in the following table.

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Examples 2 and 3 and Comparative Example 2

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The same procedure as that of Example 1 was repeated except that the silicone oil used as the component (B) was replaced with fluorocarbon oil or polytetrafluoroethylene (PTFE) and that the silicon-containing polyacetal copolymer prepared in Referential Example 4 was used as the component (C). The dispersibility was excellent and the bleeding on the surface was scarcely observed. Other results are shown in the following table.

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The same procedure as that of Example 3 was repeated except that the composition free of the silicon-containing polyacetal copolymer was used. The results are shown in the following table as those of Comparative Example 2.

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Examples 4 to 6 and Comparative Example 3

The same procedure as above was repeated except that polybutylene terephthalate (PBT) was used as the base polymer, i.e. component (A), the same substance as that used in Example 1, 2 or 3 was used as the component (B) and the silicon-containing polyacetal copolymer prepared in Referential Example 5 was used as the component (C) [in Comparative Example 3, the same procedure as in Example 4 was repeated except that no component (C) was used]. As for the effects of them, the homogeneous dispersion could be obtained in all the cases excluding that of Comparative Example 3. Other results are shown in the following table.

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Example 7 and Comparative Example 4

An epoxy resin hardener (Tohmide#255; a product of Fuji Kasei Co.) as the component (A) was heated to melt together with the silicon-containing polyacetal copolymer prepared in Referential Example 4 as the component (C) to form a homogeneous dispersion, which was left to cool. An epoxy resin (Epikote #828; a product of Yuka Shell Co.) as the component (A) and a silicone oil (Silicone Oil SH-200; a product of Toray Silicone Co.) as the component (B) were added to the dispersion and they were thoroughly mixed. The mixture was cast and hardened at 100 ° C for 3 h. The characteristic values of the hardened product are also shown in the following table.

The epoxy resin was hardened under the same conditions as in Example 4 except that the silicon-containing polyacetal copolymer as the component (C) was not used and the characteristic values thereof were determined. The results are shown in the following table.

Table Examples and Comparative Examples

	A (parts by wt.)	B (parts by wt.)	Silicon-containing copolymer (C) (parts by wt.)	Coefficient of dynamic friction	Bending test (Note-1)	Amount of oil extracted with solvent (§) (Note-2)
Ex. 1	POM " (94.5) " (95.5) " (95.5)	silicone oil (4) " (4) " (4)	Ref. Ex. 1 (1.5) Ref. Ex. 2 (0.5) Ref. Ex. 3 (0.5)	0.07 0.08 0.07	O O O	0.8 0.9 0.7
Comp. Ex. 1	POM (96.0)	silicone oil (4)	-	0.13	x	36.8
Ex. 2	POM (95.5)	fluorocarbon oil (4)	Ref. Ex. 4 (0.5)	0.07	O	2.1
Ex. 3	POM (91.0)	PTFE (8)	Ref. Ex. 4 (1.0)	0.08	Δ	-
Comp. Ex. 2	POM (92.0)	PTFE (8)	-	0.09	x	-
Ex. 4	PBT (91.5)	silicone oil (8)	Ref. Ex. 5 (0.5)	0.13	O	0.9
Ex. 5	PBT (91.5)	fluorocarbon oil (8)	Ref. Ex. 5 (0.5)	0.11	O	0.7
Ex. 6	PBT (91.5)	PTFE (8)	Ref. Ex. 5 (0.5)	0.13	O	1.0
Comp. Ex. 3	PBT (92.0)	silicone oil (8)	-	0.17	x	27.4
Ex. 7	Epikote 828 Tohmide (67.8) 255 (23.7)	silicone oil (8)	Ref. Ex. 4 (0.5)	0.16	Δ	1.1
Comp. Ex. 4	Epikote 828 (67.8) 255 Tohmide (23.7)	silicone oil (8)	-	0.24	x	17.9

Note 1 The results of the bending tests were classified into three ranks by the visual observation of delamination on the bent surface.

O: No delamination on the surface. Δ: The surface was peeled and cracks were formed.

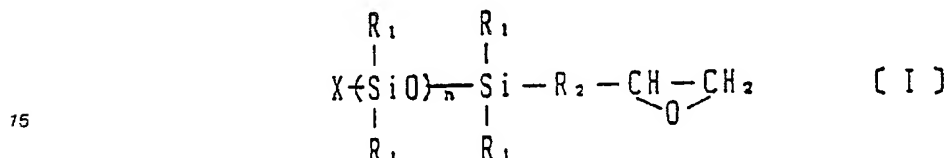
x: Breakage was caused in 24 times bending test or a part of the test piece was broken.

Note 2 Amount of extracted oil (§) based on the component (B) used.

Claims

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1. A thermoplastic resin composition comprising a thermoplastic resin (A) having a solubility parameter δ of at least $9.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ in which a modifier (B) having a solubility parameter δ of not higher than $8.5 \text{ cal}^{1/2}\text{cm}^{-3/2}$ is dispersed to form a dispersion, characterized by containing a dispersibility improver (C) comprising a silicon-containing polyacetal copolymer prepared by bonding 0.01 to 30 wt % of a terminal-reactive silicone oil of the following general formula [1]:



wherein R₁ represents an alkyl or phenyl group, R₂ represents a divalent organic group, X represents a group selected from the group consisting of -NH₂, -OH, -R₃CH-CH₂, -R₄OH, -R₅COOH, -R₆COOR₇, -R₈ and -OR₉, R₃ represents a divalent organic group, R₄ to R₆ each represent an alkylene group having 1 to 10 carbon atoms, R₇ to R₉ each represent an alkyl group having 1 to 10 carbon atoms and n represents a number of 5 to 10.

with 99.99 to 70 wt.% of a polyacetal segment through a covalent bond, the degree of polymerization of the siloxane segment (n in the formula [I]) being 5 to 1,000 and the degree of polymerization of the polyacetal segment being 5 to 2,000, in which the weight ratio of (A) to (B) is 99.9 to 85/0.1 to 15 and the amount of (C) is 0.01 to 5 parts by weight for 100 parts by weight in total of (A) and (B).

2. A thermoplastic resin composition according to claim 1, wherein the thermoplastic resin (A) is one or more thermoplastic resins selected from the group consisting of polyesters, polyamides, polyesteramides, polyurethanes, polyvinyl chloride, polycarbonates, polyacetals, polyvinylidene chloride, polyacrylonitrile, polymethacrylonitrile, polymethyl acrylate, polyvinyl alcohol, cellulose and derivatives of them.

3. A thermoplastic resin composition according to claim 2, wherein the thermoplastic resin (A) is at least one polymer selected from the group consisting of polyacetals, polyesters and polyamides.

4. A thermoplastic resin composition according to claim 3, wherein the thermoplastic resin (A) is a polyacetal homopolymer or copolymer comprising polyoxymethylene units as the main skeleton.

5. A thermoplastic resin composition according to any of claims 1 to 4, wherein the modifier (B) is one or more homopolymers selected from the group consisting of polytetrafluoroethylene, polyvinylidene chloride, polychlorotrifluoroethylene, fluororubber, polydimethylsilicone, polyalkylsiloxanes, polyphenylsiloxane, silicone rubber, butyl rubber, polyisobutylene, polyethylene, ethylene/propylene rubber and polypropylene and/or a copolymer comprising at least one of the monomer units forming these polymers as the constituent.

6. A thermoplastic resin composition according to any of claims 1 to 4, wherein the modifier (B) is a fluorocarbon oil, silicone oil or mineral oil having a molecular weight of 200,000 or less.

7. A thermoplastic resin according to any preceding claim, wherein in the formula [I] for the terminal-
45 reactive silicone oil used to prepare the silicon-containing polyacetal copolymer (C), R₁ represents an alkyl
group with 1 to 4 carbon atoms, R₂ and R₃ each represent a group of the formula -R₁₀-Y-R₁₁- in which R₁₀
and R₁₁ each represent an alkyl group with 1 to 6 carbon atoms and Y represents oxygen or sulphur, or R₂
and R₃ each represent an alkylene group with 1 to 12 carbon atoms or an oxyalkylene group with 1 to 12
carbon atoms.

50 8. A thermoplastic resin according to claim 7, wherein in the formula [I] for the terminal-active silicone oil used to prepare the silicon-containing polyacetal copolymer (C), R₁ represents a methyl group and R₂ and R₃ each represent a -CH₂-, -OCH₂-, -CH₂CH₂CH₂OCH₂- group which has a degree of polymerisation of 5 to 1000.

9. A thermoplastic resin according to any preceding claim, wherein in the silicon-containing polyacetal 55 copolymer(C) the weight ratio of the terminal-reactive silicone oil to the polyacetal segment is 0.01 to 30.99.99 to 70.

10. A thermoplastic resin according to any preceding claim, wherein the amount of the silicon-containing polyacetal copolymer(C) is 0.1 to 2 parts by weight for 100 parts by weight in total of the thermoplastic resin (A) and the modifier (B)

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